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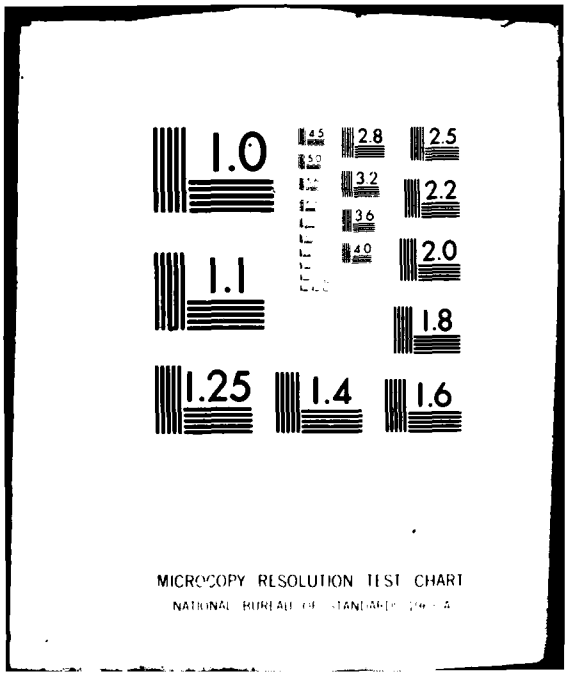
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# RAMAN-INDUCED KERR EFFECT STUDIES

FINAL REPORT

Robert W. Hellwarth

7/15/76-7/14/79

U. S. ARMY RESEARCH OFFICE

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The Raman-Induced Kerr Effect, developed at U.S.C., and the subject of this project, is a new form of laser spectroscopy which is beginning to find wide application as a remote diagnostic sensor of fast (picosecond to nanosecond) transient shocks, explosions, and plasma arcs.

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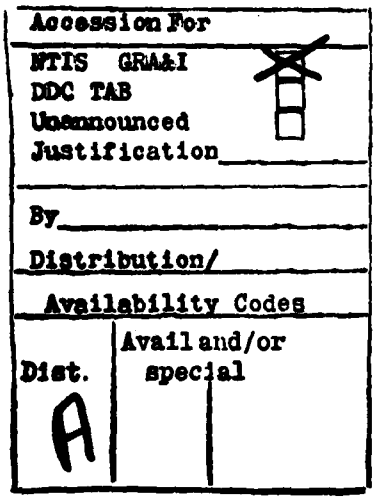
## 1. Foreword

The Raman-induced Kerr Effect, developed at U.S.C. and the subject of this project, is a new form of laser spectroscopy which is beginning to find wide application as a remote diagnostic sensor of fast (picosecond to nanosecond) transient shocks, explosions, and plasma arcs.



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## 1. Objectives

This project aims to explore the capabilities of a new laser-spectroscopic technique devised in this laboratory, by both experimental and theoretical investigations. The technique is based on monitoring the changes in optical Kerr effect which occur when the frequencies of the two ("pump" and "pulse") beams differ by the frequency of an excitation in the medium.[12] Hence, we have called the technique "Raman-induced Kerr effect" or RIKE. One special capability of RIKE which we have demonstrated is the ability to record an entire Raman spectrum of a liquid in a few nanoseconds ($\sim 10^{-8}$ sec), a spectrum which takes minutes to record in a standard Raman spectrometer.

## 2. Main accomplishments (7/15/76 to 7/14/79)

The main accomplishments of this project have been reported in the five previous reports submitted on this project.

The first year was devoted to equipment building and to extensive pulse RIKE measurements on liquids at small frequency shifts which yielded anomolous and scattered results which were apparently due to uncontrollable electrostrictive effects. Time-delayed RIKE measurements with 80 psc pulses in glycerol failed to reveal the delayed effect which was anticipated to

exist if the induced molecular alignment decayed with time constant (~500psc) associated with the high viscosity of the liquid. (See the first annual report for details.)

An initial aim of this project was to explore RIKE as a diagnostic tool for very fast transient media such as explosions, sparks, and plasma arcs.[3] Since we had no special equipment for producing these transient media, we had hoped to achieve these studies in collaboration with other labs. To this end we have initiated collaboration with Dr. Nicole Peacock of Culham Laboratory, England, who operates a Z-pinch plasma machine of appropriate parameters. He also has access to a ten joule Nd: glass laser a part of whose beam pumps a tunable dye laser. These lasers, according to theory done on this project, should produce observable RIKE in the plasma. From the shape of the RIKE spectrum we expect to be able to deduce the plasma density and the electron and ion temperatures as a function of time, as the plasma evolves. Mr. Richard Kirk, a graduate student at Royal Holloway College, is working under Peacock's supervision on this project. Mr. Kirk hopes to submit the results in partial fulfillment with requirements for a Ph.D. degree.

A second collaboration is underway with Professor Benjamin Lax at the National Magnet Laboratory (M.I.T.) to diagnose the plasma produced by the ALCATOR plasma machine there.

A third collaboration was begun with Dr. Charles Christoe of the Army Ordinance Research Laboratory at Picatinny, N.J. Dr. Christoe visited and worked in our U.S.C. laboratory from September '77 to January '78 with the intention of reproducing our RIKE apparatus in his laboratory where he could study chemical explosions in existing apparatus there. Unfortunately, his laboratory was phased out before his efforts could come to fruition.

An extension of RIKE spectroscopy to optical waveguides was conceived and studied theoretically. This should enable study of sub-microliter samples, i.e., much smaller samples than are required for free-beam spectroscopy. This work is included in a publication,[4] a reprint of which is included here as an Appendix.

While our own experimental studies attempting to obtain complete spectra with single nanosecond (or picosecond) laser pulses have not yet succeeded in highly transient media, experiments elsewhere in the U.S.C. Quantum Electronics Laboratory have succeeded in developing RIKE to equal or excell the sensitivity of all other laser-Raman techniques (CARS, stimulated gain spectroscopy, etc.) for detecting low densities of molecules in vapor or solution in the quiescent state.[5,6] Similar applications of RIKE have been inaugurated at other laboratories. In summary, we feel that the contributions of this project to the new form of laser spectroscopy called Raman-induced Kerr effect (RIKE) will be.

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# APPENDIX

6. Appendix 1

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# Optically Heterodyned Coherent Raman Spectroscopy

GARY L. EESLEY, M. D. LEVENSON, AND WILLIAM M. TOLLES

*Abstract*—A convenient variation of Raman-induced Kerr effect spectroscopy (RIKES) which overcomes many of the previous limitations of coherent Raman spectroscopy is introduced. Quantum limited signal-to-noise ratios may be approached by the use of optical heterodyne detection (OHD). Detector current, linearly proportional and phase sensitive to the third-order nonlinear susceptibility, and linearly proportional to the Raman scattering cross section, is produced. Heterodyne detection in coherent Raman spectroscopy enables the detection of weak Raman signals that would otherwise be obscured by noise resulting from background sources.

THE spectroscopic applications of a number of four-photon parametric mixing processes have recently drawn widespread attention. The main advantage of these techniques for Raman spectroscopy lies in the fact that the signals detected are orders of magnitude stronger than those found by spontaneous scattering and that the Raman spectrum can then be obtained even in the presence of strong luminescent backgrounds. The improved signal levels obtained do not, however, directly translate into noise-free spectra. The spectroscopic information obtained by all of the present techniques of coherent Raman spectroscopy is degraded by the presence of noise resulting from processes generally unimportant in spontaneous scattering. In particular, coherent anti-Stokes Raman spectroscopy (CARS) is sensitive to noise resulting from fluctuations in the power and mode structure of the input lasers. A nonresonant signal is always produced by the CARS four-photon interaction, and variations in this background signal easily obscure structures due to weak Raman modes [1]. Other techniques which suppress this background either introduce other signals equally as troublesome or unacceptably suppress the desired Raman signals. Successful application of all of these techniques requires a considerable level of experimental sophistication [2].

We report here a convenient new variation of Raman-induced Kerr effect spectroscopy (RIKES) which overcomes many of the previous limitations of RIKES and the other forms of coherent Raman spectroscopy [3], [4]. Large improvements in the signal-to-noise ratio result from the use of optical heterodyne detection (OHD) to extract electrical signals linearly proportional to the nonlinear susceptibility and linearly proportional to the spontaneous Raman cross section. The stability requirements upon the input lasers are reduced, and background levels are completely suppressed far from resonance conditions. The application of heterodyne detection transforms the RIKE into a practical tool for the detection of weak Raman modes of luminescent samples.

In the RIKE, a circularly or linearly polarized pump laser at $\omega_1$ induces an intensity-dependent birefringence in a Raman active sample. This birefringence alters the state of polarization of a second probe wave at $\omega_2$. These phenomena can be described in terms of a third-order nonlinear optical susceptibility which shows dispersive and resonant behavior when $\omega_1 - \omega_2$ approaches the frequency of a Raman mode.

If the probe wave is initially linearly polarized in the $x$ direction, the effect of the birefringence is to produce a component polarized in the $y$ direction

$$E_R(\omega_2) = E_y(\omega_2) = \frac{24\pi i\omega_2 l_{\text{eff}}}{cn_2} \chi^{(3)}_{\text{eff}}(-\omega_2, \omega_1, -\omega_1, \omega_2) E_x(\omega_1) E_y^*(\omega_1) E_x(\omega_2). \quad (1)$$

In (1), $E_x(\omega_1)$ and $E_y^*(\omega_1)$ are the complex amplitudes of the $x$ and $y$ components of the pump wave, $E_x(\omega_2)$ is the probe wave amplitude, $n_2$ is the refractive index at $\omega_2$, and $l_{\text{eff}}$ is the effective interaction length of the two beams. For a pump beam linearly polarized at 45° in the $xy$ plane, $E_x(\omega_1) = \pm E_y(\omega_1)$ and the effective nonlinear susceptibility is

$$\chi^{(3)}_{\text{eff}} = \chi^{(3)}_{2211}(-\omega_2, \omega_1, -\omega_1, \omega_2) + \chi^{(3)}_{2121}(-\omega_2, \omega_1, -\omega_1, \omega_2) = \{2\gamma + 2B(0) + B(\omega_1 - \omega_2) + 2A(\omega_1 - \omega_2)\}/24 \quad (2)$$

while for a circularly polarized pump $E_x(\omega_1) = \pm iE_y(\omega_1)$ and

$$\chi^{(3)}_{\text{eff}} = \{\chi^{(3)}_{2211}(-\omega_2, \omega_1, -\omega_1, \omega_2) - \chi^{(3)}_{2121}(-\omega_2, \omega_1, -\omega_1, \omega_2)\} = \{B(\omega_1 - \omega_2) - 2A(\omega_1 - \omega_2)\}/24. \quad (3)$$

In (2) and (3), $A(\Delta\omega)$ and $B(\Delta\omega)$ are the complex nuclear response functions which can be related to the cross sections for polarized and depolarized Raman scattering $\sigma_{xx}$ and $\sigma_{xy}$, at frequency shift $\Delta\omega$ [3]

$$\text{Im } A(\Delta\omega) = \frac{\pi c^4}{\hbar\omega_1\omega_2^3} \left\{ \frac{d^2}{d\Omega\, d\Delta\omega} \left( \frac{1}{2}\sigma_{xx} - \sigma_{xy} \right) \right\} \cdot (\exp^{\hbar\Delta\omega/kT} - 1) \quad (4)$$

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$$\text{Im } B(\Delta\omega) = \frac{\pi c^4}{\hbar\omega_1\omega_2^3} \frac{d^2\sigma_{xy}}{d\Omega\, d\Delta\omega} (\exp^{\hbar\Delta\omega/kT} - 1). \quad (5)$$

The parameter $\gamma$ describes the intrinsic electronic nonlinearity which gives rise to the background signals in CARS, while $B(0)$ reflects orientational contributions to the nonlinear susceptibility.

Ordinarily, the intensity of the $y$ polarized component of the RIKES output $I_R = (n_2c/8\pi)|E_y(\omega_2)|^2$ is detected photographically or photoelectrically. When a circularly polarized pump is used, this intensity only appears as the result of Raman resonances. The intensity can be quite large; however, it is proportional to the square of the Raman scattering cross section and therefore weak modes are disproportionately more difficult to detect. Moreover, static birefringence in the sample and optics will produce a spurious background intensity in the signal polarization condition. As in the case of CARS, noise resulting from fluctuations in laser intensity can cause changes in the background level which mask weak Raman modes. Optical means can be employed to eliminate the depolarized component due to birefringence that is uniform over the area of the probe beam [5]. Other effects, particularly small nonuniform strain fields and forward scattering, lead to a residual background intensity $I_B$ which adds incoherently to the intensity due to the RIKES interaction. The total intensity in the detected polarization is then $I_T = I_R + I_B$.

In our heterodyne detection technique, a local oscillator field $E_{LO}(\omega_2)$ polarized in the $y$ direction is purposely inserted at the detector. The local oscillator field adds coherently to the RIKES field producing a total intensity at the detector

$$I_T = \frac{n_2c}{8\pi}|E_{LO} + E_R|^2 + I_B$$

$$= I_{LO} + I_B + I_R + \frac{n_2c}{8\pi}(E_R^*E_{LO} + E_{LO}^*E_R). \quad (6)$$

If $I_{LO} \gg I_R$, the cross term $H = \frac{n_2c}{8\pi}(E_R^*E_{LO} + E_{LO}^*E_R)$ is larger than the former RIKES intensity; it is also linear in the scattering cross section. The heterodyne term $H$ can be extracted electronically from the other contributions to the total intensity at the detector. Spectra which plot the magnitude of $H$ as a function of $\omega_1 - \omega_2$ should show reduced noise levels in comparison with spectra taken by more conventional CARS or RIKES techniques.

The relative phase of the RIKE and local oscillator field is an important parameter in describing the spectrum. Equation (1) relates the phase of $E_y(\omega_2)$ to that of the probe wave $E_x(\omega_2)$. If the local oscillator field at the detector is in phase with the probe wave $E_{LO} = \eta E_x(\omega_2)$, and the pump is circularly polarized,

$$H = 192\pi^2\eta\omega_2 l_{\text{eff}} \frac{I(\omega_1)I_x(\omega_2)}{n_1n_2c^2} \text{Re}\{\chi^{(3)}_{\text{eff}}\}. \quad (7)$$

This situation can be obtained by rotating the analyzer which defines the detected polarization away from the $y$ axis. If $\theta$ is the angle of rotation $\eta = \sin\theta \approx \theta$. Alternatively, a retardation plate can be inserted in the probe beam to create a local oscillator 90° out of phase with $E_x(\omega_2)$. If $E_{LO} = i\eta E_x(\omega_2)$,

$$H = 192\pi^2\eta\omega_2 l_{\text{eff}} \frac{I(\omega_1)I_x(\omega_2)}{n_1n_2c^2} \text{Im}\{\chi^{(3)}_{\text{eff}}\} \quad (8)$$

and the heterodyne detected signal reproduces the spontaneous scattering spectrum. For an isolated Lorentzian Raman mode, $H = 2(I_RI_{LO})^{1/2}$ in this polarization condition. If the pump beam is linearly polarized at 45° in the $xy$ plane, (7) and (8) interchange.

A formal signal-to-noise analysis begins with an expression for the detector current produced by the illuminating intensities [6]

$$i(t) = G\frac{qeA}{\hbar\omega_2}[I_{LO}(t) + I_B(t) + I_R(t) + H(t)]. \quad (9)$$

The detector gain, area, and quantum efficiency are denoted respectively by $G$, $A$, and $q$. The photon energy is $\hbar\omega_2$, and $e$ is the electronic charge.

The dominant signal current produced by the OHD technique is given by the last term of (9):

$$i_H(t) = G\frac{qeA}{\hbar\omega_2} H(t) \quad (10)$$

where the time dependence of $H$ results from the particular time dependence of the pump and probe laser intensities as seen in (7) and (8). The mean-square signal current is given by

$$\langle i_H^2\rangle = \left[\frac{qeAG}{\hbar\omega_2} H\right]^2 D_H \quad (11)$$

where $D_H$ represents an effective signal "duty-factor" resulting from a particular signal processing scheme.

If the signal processing electronics integrates the signal current over an aperture time $T$, and if the time response of the current $i_H(t)$ has a maximum at $t = 0$, then

$$D_H = \frac{1}{T}\int_{-T/2}^{T/2} [i_H(t)/i_H(0)]^2\, dt. \quad (12)$$

Minimum electrical noise is generated in such a process by maintaining an electronic bandwidth-aperture-time product of unity; i.e.,

$$\Delta\nu T = 1$$

where $\Delta\nu$ is the bandwidth of the processing system. It is also advantageous to match the aperture time to laser pulselengths if a pulsed system is used.

The system noise currents arise from three basic sources: 1) shot-noise fluctuations; 2) thermal noise of the detector load; and 3) noise arising from classical laser power fluctuations. The shot noise which results from the quantized nature of detection is given by the mean-square current

$$\langle i_s^2\rangle = 2eG^2\left[i_d + \frac{qeA}{\hbar\omega_2}(I_{LO} + I_B)D_s\right]\Delta\nu \quad (13)$$

where $i_d$ is the detector dark current and $D_S$ is defined analo-

gously to (12), except that the normalized current response to $I_{LO}(t) + I_B(t)$ is substituted.

The thermal noise resulting from the detector load is given by

$$\langle i_T^2 \rangle = (4kT_e/R)\Delta\nu \tag{14}$$

where $R$ is the load resistance and $T_e$ is the "effective" temperature of the load, which accounts for noise generated in the processing electronics as well.

To account for classical fluctuations of the pump laser, we define the mean-square deviation of the pump laser power in bandwidth $\Delta\nu$ as $\epsilon_1$, giving the mean-square noise current

$$\langle i_P^2 \rangle = \epsilon_1 \langle i_H^2 \rangle. \tag{15}$$

The noise due to power fluctuations in the probe and local oscillator arising from a mean-square deviation of $\epsilon_2$ in bandwidth $\Delta\nu$ is

$$\langle i_F^2 \rangle = \epsilon_2 \left[ G \frac{qeA}{\hbar\omega_2} (I_{LO} + I_B) \right]^2 D_F \tag{16}$$

where as noted before we have assumed $E_{LO}(\omega_2) >> E_R(\omega_2) = E_y(\omega_2)$.

The total signal-to-noise power ratio at the detector output is

$$\frac{S}{N} = \frac{\langle i_H^2 \rangle}{\langle i_S^2 \rangle + \langle i_T^2 \rangle + \langle i_P^2 \rangle + \langle i_F^2 \rangle}. \tag{17}$$

Equation (17) can be expressed in a relatively simple form when $H = 2(I_R I_{LO})^{1/2}$ and the thermal noise and detector dark current can be ignored. In terms of the optical intensities at the detector, the signal-to-noise ratio is then

$$\frac{S}{N} = \frac{4D_H I_R I_{LO}/I_B}{\epsilon_2 D_F I_B [N(I_{LO}/I_B + 1) + (I_{LO}/I_B + 1)^2] + 4\epsilon_1 D_H I_R I_{LO}/I_B} \tag{18}$$

where the parameter

$$N = \frac{2\hbar\omega_2 D_S \Delta\nu}{\epsilon_2 q A I_B D_F} \tag{19}$$

describes the relative importance of the quantum and classical noise sources. The local oscillator power which maximizes this signal-to-noise ratio depends upon the parameter $N$ as

$$I_{LO} = I_B[1 + N]^{1/2}. \tag{20}$$

If the probe laser fluctuations are large enough such that $\epsilon_2 >> 2\Delta\nu\hbar\omega_2/qAI_B$, then the optimum local oscillator power will be equal to the power reaching the detector due to residual system birefringence $I_B$. However, for a well-stabilized probe, (20) suggests the use of a large local oscillator field. In the limit of perfect stabilization, (20) implies that the local oscillator should be as strong as the probe. In that case, the amplification of the local oscillator due to stimulated Raman gain makes as large a contribution to the detected signal as the RIKE itself. This amplification of the local oscillator can result in a distorted lineshape. Distorted lineshapes also result when the heterodyne signal is comparable to the ordinary RIKES signal. The range of local oscillator intensities within which the lineshapes are not distorted by such interference phenomena is $I_R << I_{LO} << I_X(\omega_2)$. Whenever (20) indicates an optimal local oscillator $\geq \sim 0.01\ I_X(\omega_2)$, the heterodyne-detected RIKES technique will prove inferior to stimulated Raman gain spectroscopy. Similarly, when (20) indicates $I_{LO} \leq \sim 100\, I_R$, intensity-detected RIKES will be necessary in order to obtain an undistorted lineshape.

The maximum signal-to-noise ratio predicted by (20) is

$$\frac{S}{N} = \frac{4I_R D_H}{\epsilon_2 I_B D_F (N + 2(N+1)^{1/2} + 2) + 4\epsilon_1 I_R D_H}. \tag{21}$$

For $N >> 10$ only the term linear in $N$ need be retained:

$$\frac{S}{N} \longrightarrow \frac{4I_R D_H}{2\hbar\omega_2 D_S \Delta\nu/q + 4\epsilon_1 I_R D_H}. \tag{22}$$

This corresponds to the case where probe fluctuations are an unimportant noise source in comparison to shot noise and pump laser fluctuations.

In the limit that both the probe and pump fluctuations are negligible,

$$\frac{S}{N} \xrightarrow[\epsilon_2 \to 0]{\epsilon_1 \to 0} \frac{2qAI_R}{\hbar\omega_2 \Delta\nu} \left( \frac{D_H}{D_S} \right) \tag{23}$$

the quantum limit. Note that while the signal for OHD-RIKES is proportional to $E_y(\omega_2)$ and thus linear in Raman cross section, the signal-to-noise ratio remains quadratic in cross section. Spontaneous scattering gives both a signal and a signal-to-noise power ratio linear in Raman cross section under similar conditions.

In contrast, the signal-to-noise ratio for intensity-detected RIKES is

$$\frac{S}{N} = \frac{I_R^2}{\epsilon_2 I_B^2 \{N[I_R/I_B + 1] + [I_R/I_B + 1]^2\} + \epsilon_3 I_R^2} \tag{24}$$

where dark current shot noise and thermal noise have been neglected and all duty factors have been set equal to one. The second term in the denominator results from pump laser fluctuations with $\epsilon_3$ being the mean-square deviation of $I^2(\omega_1)$. In the limit of negligible probe and pump fluctuations, this ratio becomes

$$\frac{S}{N} \xrightarrow[\epsilon_3 \to 0]{\epsilon_2 \to 0} \frac{qAI_R}{2\hbar\omega_2} \left[ \frac{I_R}{I_R + I_B} \right] \frac{1}{\Delta\nu}. \tag{25}$$

Comparison of this quantum-limited ratio to the heterodyne ratio in (23) reveals that intensity detection gives a signal-to-noise ratio at least four times smaller than heterodyne detection. Only when the intensity fluctuations in the pump laser constitute the dominant noise source does intensity detection give signal-to-noise ratios comparable to heterodyne detection. This situation occurs in practice only when the RIKES signal is relatively large; for weak signals, heterodyne detection always yields significant advantages.

The initial demonstrations of heterodyne RIKES were performed with pulsed lasers and described in [7] and [8]. More recent work has been completed using the hybrid CW

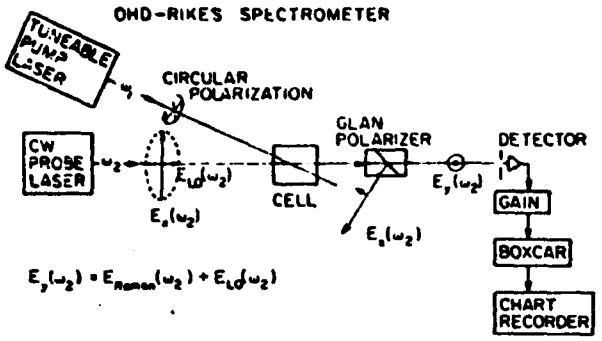


Fig. 1. Optically heterodyned RIKE spectrometer.

probe and pulsed pump laser system shown in Fig. 1. The probe laser is a 0.5-W argon-ion laser, running single mode with approximately 0.5 percent rms intensity fluctuations. The pump laser is a nitrogen pumped tunable dye laser with a peak power of 40 kW in an 8-ns pulsewidth. The detector is a photomultiplier tube with a gain of 100 and quantum efficiency of 10 percent (5145 Å), followed by an electronic wide-band amplifier (100 MHz), boxcar integrator, and chart recorder. The heterodyne signals appear on the detector output with length equal to the pulsewidth of the nitrogen laser pumped dye laser. Such pulses are easily separated from the low-frequency noise characteristic of the ion laser with the high-pass filter amplifier and the resulting signal averaged using the gated integrator. A strained glass window is inserted in the probe beam to provide a phase-shifted local oscillator, while an unshifted local oscillator is generated by rotating the glan prism polarizer.

Typical data for heterodyne as well as ordinary RIKES is shown in Fig. 2. The voltage signal-to-noise improvement provided by heterodyning is approximately a factor of 4. The dominant noise source in these experiments was RF pickup from the nitrogen laser. Improvements in equipment shielding will increase the heterodyne signal-to-noise ratio. The calculated detection limit of a similar system, utilizing a probe with 0.1 percent rms intensity fluctuations within a bandwidth of 100 MHz, is 0.001 M concentration of benzene in $CCl_4$. This represents at least one order of magnitude improvement over present coherent Raman techniques [1]. Our present system falls short of this calculated detection limit by a factor of 10 as a result of the 0.5 percent probe instability and RF noise. However, the phase-sensitive detection and improved signal-to-noise ratio are clearly demonstrated. The optical system is much simpler than that required for CARS or ASTERISK [9], and might form the basis for a simple coherent Raman spectrometer.

The great amplitude stability of CW ion and dye lasers can be exploited to improve the local oscillator stability. One strategy would be to use CW lasers for both pump and probe, and to modulate the amplitude or polarization of the pump and detect the component of the signal at this modulation frequency. In such a CW system, the duty factor would be 100 percent, and the detector bandwidth could be reduced to a few hertz. A system employing a 5000-Å probe laser stabilized to one part in $10^6$ would be quantum noise limited if $I_{pA}$ were $10^{-6}$ W, otherwise the intensity fluctuations would

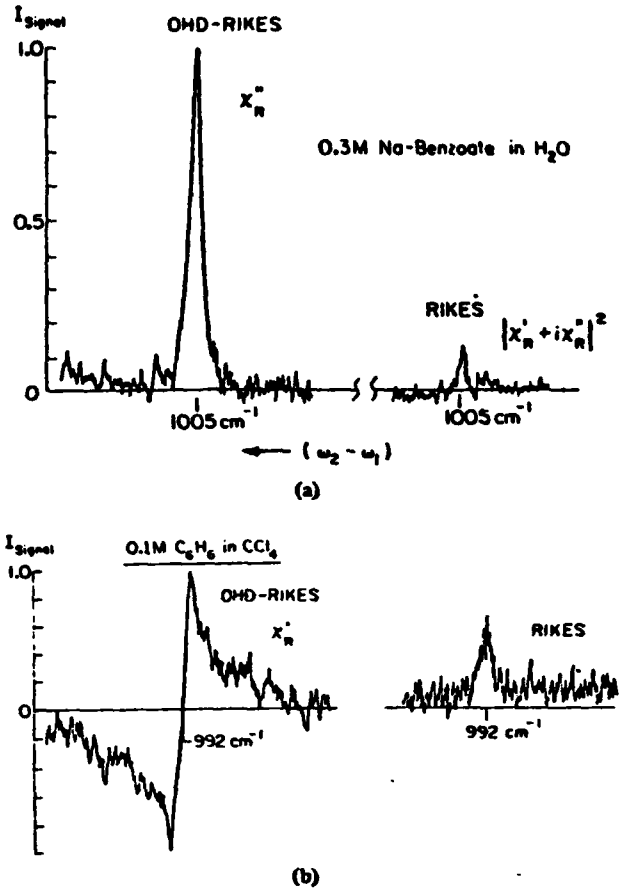


Fig. 2. Relative signal intensities derived from the 1005 $cm^{-1}$ mode of 0.3-M sodium benzoate in water, for OHD (heterodyne) as well as normal RIKES. The imaginary contribution has been selected in the heterodyned spectra. (b) Signals derived from the 992 $cm^{-1}$ mode of 0.1-M benzene in carbon tetrachloride. Real contribution is displayed in the heterodyned spectra. In both (a) and (b) the probe power was 0.5 W at 5145 Å, and the pump power was 40 kW.

still dominate. Heterodyne detection markedly improves the feasibility of CW RIKES.

It might also be noted that heterodyne detection can be employed to enhance CARS signals [10]. In this case, the nonresonant background itself supplies the local oscillator, but the frequency of one of the lasers must be modulated to facilitate detection of the cross term. In many materials, the nonresonant background is either too large or too small to give optimal local oscillator power. The output from a second sample, (preferably a material which lacks Raman modes, such as NaCl or high-pressure argon), can be added coherently to that from the Raman active material in order to optimize the local oscillator power and facilitate heterodyne detection.

The value of heterodyne detection in coherent Raman spectroscopy is that it allows the detection of Raman signals that would otherwise be buried in noise resulting from background signals. Presently available laser sources allow the quantum limit of detection sensitivity to be approached. It is particularly convenient to apply heterodyne detection to RIKES because the frequency of the RIKES signal is equal to that of one of the incident lasers. The resulting technique is both powerful and convenient; it allows the real and imaginary part

of the nonlinear susceptibility of the sample to be determined separately. When the detected signal corresponds to the imaginary part of the susceptibility, the OHD RIKES trace reproduces the spontaneous Raman scattering spectrum.

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